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11/29/89

Biologic Markers of Air-Pollution Stress and Damage in Forests

Committee on Biologic Markers of
Air-Pollution Damage in Trees

Board on Environmental Studies and Toxicology

Commission on Life Sciences

National Research Council

NATIONAL ACADEMY PRESS
Washington, D.C. 1989

MARKERS OF AIR POLLUTION IN FORESTS: NUTRIENT CYCLING

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ABSTRACT

Air pollution may affect forest nutrient cycles in a number of ways, but many of these effects are difficult to evaluate because control sites unaffected by air pollution are seldom available for comparison. Air pollution may alter either the nutrient content or the flux rates in forest ecosystems, but, with a few exceptions (e.g., SO_4^- in foliage, Al^{3+} or Pb in tree rings), flux measurements provide more sensitive measures of air pollution effects. The fact that nutrient pools are typically quite large relative to the flux rates into and out of them **accounts** for the sensitivity of flux measurements. However, nutrient pools may be significantly affected by air pollution over long periods of time, and the importance of sample archiving as a means of detecting these long-term changes in nutrient pools (e.g., soils) cannot be overemphasized. Acid deposition and/or foliar damage by ozone may cause increased foliar leaching, but the degree to which air pollution affects foliar leaching is very difficult to assess in the absence of control sites for comparison. Decomposition may also be altered by increased acid input, trace metal deposition, or gaseous pollution; but, once again, many of these potential changes are very difficult to evaluate for lack of control sites. Acid deposition nearly always influences soil leaching rates to some degree. In the case of SO_4^- , reasonable assumptions about background concentrations often allow an approximate estimate of the degree to which leaching is affected by sulfate deposition. For NO_3^- , there is considerably less certainty about exactly what background levels should be. It is therefore more difficult to evaluate the degree to which current NO_3^- leaching rates are affected by atmospheric N deposition without a full analysis of the N cycle of the ecosystem in question. The same basic considerations apply to stream concentrations. Stream concentrations may be less sensitive to air pollution effects than soil solution concentrations, but the advantages of ease of sampling and integration of whole watershed response make stream concentration monitoring an attractive **possibility as a** marker of air pollution in forest ecosystems.

INTRODUCTION

In general, nutrient fluxes (particularly solution fluxes such as foliar and soil leaching) respond most readily to air pollution, whereas nutrient pools (e.g., soil,

vegetation, and forest floor contents) are often sufficiently large to remain rather insensitive to all but very long-term effects of air pollution inputs. Even in cases where long-term changes in **nutrient** pools have occurred, they may be difficult to identify and demonstrate if no control sites unaffected (or less affected) by air pollution are available for comparison. Thus, with the exception of certain key nutrients or ions in certain ecosystem components, nutrient fluxes usually represent more sensitive markers of the presence of air pollution than do nutrient pools. The **presence** of air pollution by no means **implies** that this pollution is creating any damage to the ecosystem, however. From a nutrient cycling perspective, damage may be defined as **either** the depletion of a critical nutrient or the elevation of any element to **toxicity levels**. Thus, while nutrient or element pools are relatively insensitive markers of the presence of air pollution, they may be the most sensitive markers of pollutant damage.

In the following discussion, we review very briefly the literature regarding effects of air pollution on various nutrient fluxes and contents in forest ecosystems, attempt to extract from that literature useful markers of air pollution on forest nutrient cycles, and make some recommendations as to general sampling protocols that may be useful in monitoring pollutant presence and effects on forest nutrient cycles.

DEPOSITION AND CANOPY INTERACTIONS

There has long been a concern over the effects of acid deposition on foliar leaching. Studies involving artificial acid irrigation have variously found either no effect on leaching (e.g., Haines et al. 1985) or an increase in foliar leaching (at very low pH) (e.g., Wood and **Bormann** 1974). Amthor (1986) argues convincingly that the energy costs of increased **uptake** to compensate for accelerated foliar leaching are inconsequential. However, there may be significant nutritional consequences if uptake does not compensate for these losses. Rehfuess (1987) and Prinz (1985) advance a foliar leaching hypothesis to explain the decline of high-elevation forests in Germany. They hypothesize that foliage predamaged by exposure to **photo-oxidants** like ozone is especially susceptible to increased Ca and Mg leaching. In soils low in exchangeable Ca and Mg, uptake apparently does not compensate for this additional foliar leaching, and foliar Ca and Mg concentrations decline, eventually to deficiency levels. These deficiencies, **in** turn, lead to reduced frost hardiness, reduced chlorophyll content and photosynthesis rates, and reduced root growth and uptake.

Unfortunately, the lack of control sites presents serious problems for testing the **foliar** leaching hypothesis and for using foliar leaching as a marker of air pollution stress. As noted by Lovett et al. (1986), the disappearance or **H⁺** in the forest canopy does not necessarily mean that foliar leaching has increased: the **H⁺** **may** simply have gone into a neutralization reaction with organic **anions** that naturally leach from the canopy. This does not imply, however, that acid deposition has no effect on the ecosystem: **H⁺** in combination with organic acid would still result in **throughfall** with greater total acidity than that in **throughfall** in an unpolluted environment. Low foliar Mg could not serve **as** a reliable marker either because Mg deficiencies could clearly occur in pristine as well as in polluted environments.

Tissue concentration of certain pollutants has obvious potential use as a marker of **air** pollution. For instance, Bieberdorf et al. (1958) found that **SO₄⁻** in *Pinus taeda* foliage was a very sensitive index of proximity to an **SO₂** source, presumably because of **SO₂** absorption by the foliage. Such accumulation of **SO₄⁻** in the foliage will occur only to the extent that trees are not S deficient. Kelly and Lambert (1972) indeed found foliar **SO₄⁻** to be a reliable indicator of S excess (i.e., **SO₄⁻** accumulates only when the S

supply is greater than tree S requirements). Foliar SO_4^{2-} may not be a sensitive index of greatly excessive S deposition, **either**: foliar SO_4^{2-} may reach as high as 50% of total foliar S, but apparently does not exceed this value even in areas of **very** high S deposition (Johnson 1985).

DECOMPOSITION, NUTRIENT MINERALIZATION, AND NITRIFICATION

Most studies of the effects of air pollution on decomposition have **been** conducted under conditions of artificial irrigation, often with extremely high inputs over short time periods, raising serious questions about the applicability of these results to actual conditions. For example, would soil organisms be able to adapt to more acidic conditions over longer periods with lower inputs? Moreover, results have been inconsistent between sites and from one study to another. For **instance**, Like and Klein (1985) found that acid treatments had no effect upon nitrification but stimulated nitrogen mineralization in soil columns from Camel's Hump, Vermont, whereas **Strayer et al.** (1981) found that acid irrigation had a negative effect on nitrification and either no effect or a stimulative effect upon nitrogen mineralization of soils from Panther Lake in the **Adirondacks** in New York. Klein et al. (1983) reported that the same treatment enhanced nitrification in one soil from the Adirondacks and inhibited it in two other soils from the same region. **Stroo** and Alexander (1986) brought forward the suggestion that decreased N mineralization does not necessarily represent a decline in N availability because part of the N supply could be **entering** with acid precipitation.

Baath et al. (1980) found reduced decomposition rates (measured by the litterbag technique) following acidification of forest soils in Sweden, whereas Hovland et al. (1980) found that acid irrigation caused either increases or decreases in decomposition rates (litterbag technique) depending upon the time of measurement or the amount of acid applied. Similarly, Chang and Alexander (1984) showed that simulated acid rain applied to three northeastern soils enhanced or inhibited decomposition depending on the amount of acid applied and the amount of organic acid leached from soil samples. In another study, sulfuric acid applications to hardwood leaf packs generally stimulated the loss of litter, nutrients, and trace metals from the soil surface (Lee and Weber 1983). Hay et al. (1985) suggested that acidification of the mineral horizons of a podzol soil inhibited the transport of the major classes of soil organic components but increased the transport of nitrogenous substances. Hovland (1981), Kelly and Strickland (1984), and Johnson and Todd (1984) observed little or no effect of artificial acid irrigation upon CO_2 evolution. On the other hand, in one of the few studies that did not involve artificial irrigation, **Prescott** and Parkinson (1985) found that decomposition was greatly reduced in sites near a sour gas plant that had been emitting sulfur for over 20 years, compared to that in sites farther from the plant.

In vitro studies by Moloney et al. (1983) on the degradation of spruce-fir litter from New England showed that microbial CO_2 evolution was reduced by acidic conditions, and that it was further repressed by the presence of Pb and Zn but not by Al or Cu. From these observations it was suggested that the acidity and heavy **metals** introduced by polluted rain may adversely affect the metabolism of decomposing **microflora**.

The effect of trace metals on soil microorganism activity has been examined in a number of laboratory and field studies. Generally, extremely high trace metal concentrations, such as those **found** in the vicinity of smelters, decrease soil microorganism activity. (**Zottl** 1985). **Friedland et al.** (1986) reported that levels of Pb, Zn, and Ni below 10^3 mg/kg, levels of Cu below 10^2 mg/kg, and levels of Cd below 120 rag/kg have no measurable effects on decomposition rates. In a long-term field study at

Camel's Hump, Vermont, **Friedland et al.** (1986) found substantial increases in trace metal and organic matter concentrations of the forest floor over time. However, they suggested that current metal concentrations are not capable of significantly reducing decomposition of organic **matter**.

Hovland et al. (1980) summarize their results **by** stating: "The effect of the artificial acid rain appears to be more pronounced on the leaching of metal elements than **on the** biological activity and **the** dynamics of N and P." This statement seems appropriate not only for their particular studies but also for the majority of the artificial irrigation studies taken as a whole. It would seem that decomposition and nutrient mineralization must certainly be affected by soil acidification at some point, but this may come after the many decades of acid input usually necessary to change soil acidity. In the initial stages of acid input, however, biological processes in the soil generally do not appear to be as sensitive to acid inputs as chemical processes, namely, soil leaching.

SOIL LEACHING AND NUTRIENT EXPORT

The effects of acid deposition on **soil** leaching and stream water export have been studied extensively, **are** reasonably well understood, and have been modeled (e.g., Reuss and Johnson 1986). These effects can be estimated rather easily, even in the absence of control sites, given certain reasonable assumptions about background solution concentrations of SO_4^{2-} and NO_3^- . After the assumed background SO_4^{2-} concentrations are subtracted, a comparison of the total cation or total anion concentration accounted for by SO_4^{2-} with that accounted for by HCO_3^- and organic acids usually provides an approximation of the net effect of atmospheric S deposition on nutrient leaching from soils. **Exceptions** to this principle, such as sites where significant S-bearing minerals are present or sites where **sulfur-bearing** fertilizers have been previously applied, are usually clearly defined. The situation with respect to atmospheric N deposition and soil solution NO_3^- concentrations is more complex, because **background** NO_3^- can be quite high in certain circumstances (Van Miegroet and Cole 1984, Foster 1985), and **many** different types of ecosystem disturbances **other** than air pollution can cause an increase in soil solution NO_3^- concentration. The effect of N input on soil leaching also depends on whether NO_3^- or NH_4^+ enters the soil and on whether the latter **form** is subsequently nitrified. Further complications and uncertainties arise in cases where organic acids are the dominant natural leaching agents because organic acids are commonly estimated through the calculation of an anion deficit (Johnson et al. 1977, Cronan et al. 1978).

The effect of forest soil acidification on element leaching has been shown in soils of southern Sweden (Tyler et al. 1987). Measurements taken over decades show a decline in soil pH. This decline has increased the solubility of elements such as Mg^{2+} , Al^{3+} , Cd^+ , and Zn^+ , with consequent higher metal concentrations in soil water and increased output of these metals.

The most reliable way to ascertain the effects of atmospheric S or N deposition on soil solution SO_4^{2-} and NO_3^- concentrations is through an analysis of the S and N cycles of the ecosystems in question. Atmospheric S and N deposition in excess of biological demands almost invariably results in an increase in SO_4^{2-} and NO_3^- above background or natural levels. The SO_4^{2-} and NO_3^- **can**, in turn, leach from the soil if inorganic reactions permit it. Inorganic interactions between NO_3^- and soils **are** normally minimal, and, as a general rule, atmospheric N input in excess of biological demands leads to increased NO_3^- and associated cation leaching. Fortunately, biological demands for N are often quite high, and many forests are, in fact, **N-limited** (Cole and Rapp 1981). There are exceptions, however, in which very high rates of NO_3^- leaching occur as a direct

consequence of high rates of atmospheric N deposition (Van Breemen et al. 1982) or as a result of excessive N fixation (Van Miegroet and Cole 1984).

On the other hand, biological demands for S are less than 5% of those for N (on a molar basis) and are typically exceeded by atmospheric S inputs in polluted regions (Johnson 1985). Unlike NO_3^- , however, SO_4^{2-} can adsorb to soils (such as many in the southeastern United States) rich in Fe and Al oxides and poor in organic matter. This adsorption can reduce the potential for leaching considerably up to the point where soil adsorption sites equilibrate with incoming SO_4^{2-} (Johnson 1985).

Because solutions have to be electrically neutral, increases in SO_4^{2-} or NO_3^- soil solution concentrations lead to concurrent increases in total cation concentration. The cation composition of the solution in equilibrium with a soil is controlled by both the total anion solution concentration and the composition of the cation exchange complex. These relationships are described fairly accurately by well-known selectivity equations developed more than 50 years ago (e.g., Gapon 1933). They predict, in essence, that the concentration of a given cation in soil solution is governed by the proportion of this cation on the soil exchange complex and the total ionic strength of the soil solution. Reuss (1983) points out one very interesting aspect of these equations regarding the effects of solution concentration changes: as total ionic concentration increases, the ratios of trivalent to divalent to **monovalent** cations in solution also increase. In other words, as the solution becomes more concentrated, cation concentrations increase in the following descending order: Al^{3+} , Ca^{2+} , Mg^{2+} , K^+ , Na^+ , and H^+ . Thus, Al^{3+} concentrations in soil solution increase not only as the soil acidifies (i.e., as the proportion of Al^{3+} on the exchange complex increases) but also as the total ionic concentration of soil solution increases. These equations also imply that, compared with leaching of the other major cation nutrients, K^+ leaching from soils will be the least affected by acid deposition.

These exchange equations further suggest that the H^+ concentration increases to some extent (though not as much as Al^{3+} , Ca^{2+} , or Mg^{2+}) with any increase in total ionic concentration, even without direct H^+ input from the atmosphere. This increase in H^+ (decrease in pH) causes HCO_3^- and organic anions to protonate (becoming H_2CO_3 and uncharged organic acids), leading to a decrease in the concentrations of these natural anions. This replacement of natural, weak acid anions with atmospherically introduced, strong acid anions (SO_4^{2-} and NO_3^-) is referred to as an "anion shift." It results in a lesser acceleration of total nutrient leaching than would occur with the simple addition of SO_4^{2-} and NO_3^- to the natural leaching of HCO_3^- and organic anions. Krug and Frink (1983) argued that this anion shift can be such that no net increase in total cation leaching occurs, even though the anion composition of soil solution changes dramatically. This is an extremely unlikely, if not impossible, scenario in that it would require a H^+ concentration increase equivalent to the addition of SO_4^{2-} and NO_3^- . However, the **mitigative** effect of the anion shift can be significant in acid soils, where H^+ is a significant **cationic** solution component.

CHANGES IN SOILS

Since soil nutrient pools are usually large relative to inputs and outputs of nutrients, changes in soil **content** often occur **very** slowly and are very difficult to measure. Nonetheless, **changes** in soils have been noted in certain cases. We found evidence of modern SO_4^{2-} accumulation in soils near Oak Ridge, Tennessee, by sampling beneath and adjacent to old houses (Johnson, et al., 1981). Berden et al. (1987) provide an excellent review of the literature documenting changes in soil acidity over time as

well as supply a reasonable evaluation of the potential role of acid deposition in causing these changes. Soil acidity per se is most emphatically not a good marker of air pollution effects; soils can become extremely acid due to natural leaching and plant uptake in unpolluted forests, under either intensive management (Turner and Kelly 1977) or pristine conditions (e.g., Johnson et al. 1977, Ugolini et al. 1977).

Although soils are rather insensitive to changes, we feel **strongly** that both long-term monitoring of soil chemical properties and rigorous sampling and archival protocols are highly desirable features of any program to monitor markers of air pollution. Had such a program been implemented in the past, much of the current heated debate and often unsupported speculation concerning the effects of air pollution on soils would not exist. Furthermore, the existence of archived soil samples, with proper tests for sample storage effects, would allow measurement and **remeasurement** of currently **unmonitored** soil parameters that could be of importance in the future.

CHANGES IN STREAMS

Perhaps the most reliable integrated marker of air pollution effects on forest nutrient cycles is the long-term trend or **pattern** in stream chemistry. Watershed studies have shown that alteration of solute concentrations in streams and net nutrient losses provide a sensitive indicator of ecosystem stress (O'Neill et al. 1977, Likens et al. 1977, Swank and Douglass 1977). For example, long-term studies in streams draining hardwood forests of the southern Appalachian Mountains show the beginning of delayed response to atmospheric deposition as evidenced by increased SO_4^{2-} concentrations, reduced HCO_3^- concentrations, and changes in levels of other solutes (Swank and Waide 1988). Shifts in stream chemistry have also been related to forest insect infestations and forest harvesting practices (Swank 1986). The use of stream chemistry monitoring as an indicator of air pollution in forests has several disadvantages: (1) a potential lag in response (compared to other ecosystem **compartments** more directly exposed to pollutants), (2) a lack of long-term records needed to establish trends, and (3) an inability to distinguish **cause-and-effect** relationships (in the absence of process studies). However, stream chemistry does integrate the spatial and temporal **variability** of forest nutrient cycles and may provide the most reliable index of cumulative effects of air pollution.

CONCLUSIONS

1. The most sensitive markers of the presence of air pollution are probably the concentrations of certain ions such as SO_4^{2-} and NO_3^- in natural waters. The sensitivity of a given natural water to air pollution declines **with** the degree to which the water is **affected** by biological and chemical cycling processes within the ecosystem; thus, the following order of sensitivity is generally **found**: deposition > **throughfall** > soil solution > **streamflow**. With respect to the **ease** of monitoring, the following order is found: streamflow > wet deposition > soil solution > throughfall > dry deposition.
2. Certain key ion or element concentrations in particular ecosystem components (e.g., SO_4^{2-} in foliage, Al^{3+} in tree rings, lead in forest floor) may also be useful markers of the **presence** of air pollution.

3. The mere presence of air pollution in a forest ecosystem can by no means be taken as evidence that air pollution is causing any significant damage to the ecosystem.
4. Nutrient pools are most likely to be affected by air pollution. However, due to their relatively large size, they are rather insensitive markers.
5. Sample archiving from nutrient pools of ecosystems with well-documented long-term element budgets is essential for determining long-term changes in ecosystem nutrient pools caused by air pollution as well as by other factors.

ACKNOWLEDGMENT

Research supported by the Electric Power Research Institute under Contract No. RP-2621 with Martin Marietta Energy Systems, Inc., under Contract No. DE-AC05-84OR21400 with the U.S. Department of Energy. Publication No. 3133, Environmental Sciences Division.

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